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International Application No. PCT/EP00/09789

Attorney Docket No. MG-2280

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

INTERNATIONAL APPL. NO.: PCT/EP00/09789 :

INTERNATIONAL FILING DATE: OCTOBER 6, 2000 :

PRIORITY DATE: OCTOBER 13, 1999 :

APPLICANT: GERHARD GROSS, ET AL. : ART UNIT:

SERIAL NO.: To be assigned : EXAMINER:

FILED: To be assigned :

FOR: METHOD AND APPARATUS FOR INTRODUCING POWDERED RAW MATERIAL INTO A FLUIDIZED PARTICLE BED :

Hon. Commissioner of Patents and Trademarks
Box PCT
Washington, D.C. 20231

I hereby certify that this paper, along with any other paper or fee referred to in this paper as being transmitted herewith, is being deposited with the Assistant Commissioner for Patents, Box PCT, Washington, D.C. 20231 via Express Mail on June 11, 2001 by the below indicated person.

HAROLD PEZZNER

(Typed or printed name of person mailing paper or fee)

Harold Pezzner

(Signature of person mailing paper or fee)

TRANSMITTAL OF APPLICATION PAPERS TO U.S. DESIGNATED\ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. § 371 (37 CFR 1.494 OR 1.495)

This transmittal letter is based upon PTO Form 1390.

The above-identified applicant has filed an International Application under the P.C.T. and hereby submit(s) to the United States Designated/Elected Office (DO/EO/US) the following items and other information.

1. [] This is the FIRST submission of items concerning a filing under 35 U.S.C. §371

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2. [] This is the SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. §371.

3. [] This is an express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay.

4. [x] A proper Demand for International Preliminary Examination (IPE) was made to the appropriate Authority (IPEA) by the 19th month from the earliest claimed priority date.

5. [X] A copy of the International Application as amended (35 U.S.C. §371(c)(2)) -
   (a) [X] is transmitted herewith (required when not transmitted by International Bureau).
   (b) [] has been transmitted by the International Bureau.
   (c) [] is not required, as the application was filed in the United States Receiving Office (RO/US).

6. [x] A (verified) translation of the International Application into the English language is enclosed.

7. [x] Amendments to the (specification and) claims of the International Application under PCT Article 19 (35 U.S.C. §371 (c)(3))
   (a) [x] are transmitted herewith (required if not transmitted by the International Bureau).
   (b) [] have been transmitted by the International Bureau.
   (c) [ ] have not been made; however, the time limit for making such amendments has NOT expired
   (d) [] have not been made and will not be made.
   (e) [] will be submitted with the appropriate surcharge.

8. [] A translation of the amendments to the claims (and/or the specification) under PCT Article 19 (35 U.S.C. §173(c)(3)) is enclosed or will be submitted with the appropriate surcharge.

9. [ ] An Oath or Declaration/Power of Attorney of the inventor(s) (35 U.S.C. §371(c)(4)) is enclosed.

10. [] A translation of at least the Annexes to the IPE Report under PCT Article 36 (35 U.S.C. §371(c)(5)) is enclosed.

Items 11. to 16. below concern other document(s) or information included:

11. [] An Information Disclosure Statement under 37 CFR 1.97 and 1.98 is enclosed.

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12. [] An Assignment is enclosed for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3 31. is included.

13. [x] A FIRST preliminary amendment is enclosed. **IT IS REQUESTED THAT THE FILING FEES FOR THE CLAIMS BE CALCULATED AFTER THE CLAIM AMENDMENTS IN THE PRELIMINARY AMENDMENT HAVE BEEN ENTERED.**

14. [] A substitute specification (including claims, abstract, drawing) is enclosed.

15. [] A change of Power of Attorney and/or address letter is enclosed.

16. [x] Other items of information:

[] This application is being filed pursuant to 37 CFR 1.494(c) or 1.495(c), and any **missing parts** will be filed before expiration of -

[] 22 months from the priority date under 37 CFR 1.494(c), or

[x] 32 months from the priority date under 37 CFR 1.495(c).

[x] The undersigned attorney is authorized by the International application and by the inventors to enter the **National Phase** pursuant to 37 CFR 1.494(c) or 1.495(c).

The following additional information relates to the International Application:

[x] Receiving Office: EPO (originally filed in the German Patent Office)
[] IPEA (if filing under 37 CFR 1.495): EPO
[x] Priority claim(s) (35 USC §§ 119, 365): App. No. 199 49 193.3
Filed: OCTOBER 13, 1999

[x] A copy of the International Search Report is
[x] enclosed.
[] attached to the copy of the International Application.
[] A copy of the Receiving Office Request Form is enclosed.

17. [] Small Entity Form

The fee calculation is set forth below.

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FEE CALCULATION

NOTE: IT IS REQUESTED THAT THE FILING FEES FOR THE CLAIMS BE CALCULATED AFTER THE CLAIM AMENDMENTS IN THE PRELIMINARY AMENDMENT HAVE BEEN ENTERED.

[x] A check in payment of the filing fee, calculated as follows, is attached (37 CFR 1.492)

Basic Fee	$710.00
Total Number of claims in excess of (20) times $18	$
Number of independent claims in excess of (3) times $80	$
Fee for multiple dependent claims $270	$
TOTAL FILING FEE	$710.00

Kindly send us the official filing receipt.

The Commissioner is hereby authorized to charge any additional fees which may be required or to credit any overpayment to Deposit Account No. 03-2775. This is a "general authorization" under 37 CFR 1.25(b), except that no automatic debit of the issue upon allowance is authorized. This letter is being submitted in triplicate.

Respectfully submitted,

By Harold Pezzner

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MG-2280

I hereby certify that this paper, along with any other paper or fee referred to in this paper as being transmitted herewith, is being deposited with the United States Postal Service with sufficient postage as First-Class Mail in an envelope addressed to: Commissioner of Patents and Trademarks, Washington, D.C. 20231 on this ________ day of __________________________, 2001.

By: ______________________________

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

MESSER GRIESHEIM GMBH

SERIAL NO.:

INTERNATIONAL PCT APPL. NO: PCT/EP00/09789

INT'L FILING DATE: 10/06/00

FOR: METHOD AND APPARATUS FOR INTRODUCING POWDERED RAW MATERIAL INTO A FLUIDIZED PARTICLE BED

Hon. Commissioner of Patents
and Trademarks
BOX PCT
Washington, D.C. 20231

PRELIMINARY AMENDMENT FILED CONCURRENTLY
<u>WITH FILING OF APPLICATION</u>

Sir:

Please amend the above identified application as follows:

IN THE CLAIMS:

Amend claims 4-7, 9-13, 16 and 18 as indicated in the attached clean copy of the claims. Also attached is a marked copy showing the changes.

Add the following claims:

19. The reactor according to Claim 14, characterized in that the diffuser (2) has a gas-outlet opening that is in functional connection with a pressure-reducing device (6;7).

20. The process according to Claim 2, characterized in that the distance between the supersonic nozzle (4) and the diffuser (2) is set in such a way that the pressure in the suction chamber (3) is minimal.

REMARKS:

This amendment is for the purpose of placing the claims in accord with U.S. practice.

Respectfully Submitted,
CONNOLLY BOVE LODGE & HUTZ LLP

BY: Harold Pezzner

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MARKED COPY OF THE CLAIMS

4. (amended) The process according to [one of the preceding claims] <u>Claim 1</u>, characterized in that the fluidizing gas is used as the propellant gas.

5. (amended) The process according to [one of the preceding claims] <u>Claim 1</u>, characterized in that the propellant gas contains chlorine and/or oxygen.

6. (amended) The process according to [one of the preceding claims] <u>Claim 1</u>, characterized in that the propellant gas is accelerated to a supersonic speed corresponding to at least 1.2 Mach, preferably 1.3 Mach to 3 Mach.

7. (amended) The process according to [one of the preceding claims] <u>Claim 1</u>, characterized in that the diffuser (2) has a gas-outlet opening and in that a zone of negative pressure is created in the area of the gas-outlet opening.

9. (amended) The process according to [one of the preceding claims] <u>Claim 1</u>, characterized in that the raw material dust discharged from the fluidized particle layer is used as the solids pow-der.

10. (amended) The process according to [one of the preceding claims] <u>Claim 1</u>, characterized in that primary particles are used as the solids powder.

11. (amended) The process according to [one of the preceding claims] <u>Claim 1</u>, characterized in that waste dust is used as the solids powder.

12. (amended) The process according to [one of the preceding claims] Claim 1, characterized in that the propellant-gas stream (11) is blown into the lower half, preferably into the lower quarter, of the fluidized particle layer.

13. (amended) The process according to Claim [2 or] 3, characterized in that the distance between the supersonic nozzle (4) and the diffuser (2) is set in such a way that the pressure in the suction chamber (3) is minimal.

16. (amended) The reactor according to Claim [14 or] 15, characterized in that the diffuser (2) has a gas-outlet opening that is in functional connection with a pressure-reducing device (6; 7).

18. (amended) The reactor according to [one of Claims] Claim 14 [through 17], characterized in that the supersonic nozzle (4) can be moved in the direction indicated by (9) towards the diffuser (2) and in the opposite direction.

CLEAN COPY OF THE CLAIMS

4. The process according to Claim 1, characterized in that the fluidizing gas is used as the propellant gas.

5. The process according to Claim 1, characterized in that the propellant gas contains chlorine and/or oxygen.

6. The process according to Claim 1, characterized in that the propellant gas is accelerated to a supersonic speed corresponding to at least 1.2 Mach, preferably 1.3 Mach to 3 Mach.

7. The process according to Claim 1, characterized in that the diffuser (2) has a gas-outlet opening and in that a zone of negative pressure is created in the area of the gas-outlet opening.

9. The process according to Claim 1, characterized in that the raw material dust discharged from the fluidized particle layer is used as the solids pow-der.

10. The process according to Claim 1, characterized in that primary particles are used as the solids powder.

11. The process according to Claim 1, characterized in that waste dust is used as the solids powder.

12. The process according to Claim 1, characterized in that the propellant-gas stream (11) is blown into

the lower half, preferably into the lower quarter, of the fluidized particle layer.

13. The process according to Claim 3, characterized in that the distance between the supersonic nozzle (4) and the diffuser (2) is set in such a way that the pressure in the suction chamber (3) is minimal.

16. The reactor according to Claim 15, characterized in that the diffuser (2) has a gas-outlet opening that is in functional connection with a pressure-reducing device (6; 7).

18. The reactor according to Claim 14, characterized in that the supersonic nozzle (4) can be moved in the direction indicated by (9) towards the diffuser (2) and in the opposite direction.

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Process and device to carry out a reaction between gaseous and solid reactants in a fluidized particle layer

The invention relates to a process to carry out a reaction between gaseous and solid reactants in a fluidized particle layer, whereby a fluidizing gas flows through a loose bed of primary particles, as a result of which the bed rises while forming the fluidized particle layer and then reacts with the primary particles, and whereby a stream of a propellant gas loaded with solids powder and accelerated to supersonic speed by means of a supersonic nozzle is blown into the fluidized particle layer transversally to the main flow direction of the fluidizing gas.

Furthermore, the invention relates to a reactor in which to carry out a reaction in a fluidized particle layer, said reactor having a feed tray through which a fluidizing gas is fed into a bed of primary particles located above the inflow tray in order to generate the fluidized particle layer, and having a reactor wall that surrounds the fluidized particle layer, into which wall at least one feed device is inserted above the inflow tray, said device comprising a supersonic nozzle by means of which a propellant gas is accelerated to supersonic speed, thereby generating a propellant-gas stream directed transversally to the main flow direction of the fluidizing gas.

In particular, the subject matter of the invention is a process to produce metal chlorides such as $TiCl_4$, $ZrCl_4$ and $SiCl_4$.

In order to produce such metal chlorides, granular oxidic raw materials and granular petroleum coke are reacted at temperatures above 800°C [1472°F], normally in fluidized-

bed reactors, with the fluidizing gases chlorine and oxygen so as to form metal chlorides, CO_2 and CO.

Approximately 5% to 15% of the solid reactants are discharged as dust together with the reaction gases once the grain size has been markedly reduced by the reactions. These losses of raw material owing to dust discharge significantly raise the production costs of the metal chlorides.

Up until now, the approach of returning this dust to the fluidized-bed reactors in order to achieve a complete reaction has led to unsatisfactory results.
On the one hand, it is difficult to feed in the dusts against the elevated pressure that prevails in the fluidized particle layer in the fluidized-bed reactor and, on the other hand, if the dust is fed in downstream from the feed opening, due to the slight cross mixing that is typical of fluidized-bed reactors, the result is an insufficient supply of reaction gases in comparison to the supply of highly reactive fine particles, so that a large portion of the dust is discharged once again.

The proposal has been made to pelletize the dust, but this entails considerable effort and any successful outcome is strongly dependent on the strength of the calcined pellets.

Figure 5 shows a feed device that is used to feed a solid into a reactor and that is in the form of an injector, of the type known from pneumatic conveying technology. Such injectors consist of a suction chamber (53) with a solids feed opening (55), a usually conical nozzle (51) and a diffuser (52) arranged coaxially thereto. These injectors can be employed to convey fine-particle solids from a chamber having a lower pressure and located above the feed opening (55) into a chamber having a higher pressure (reactor) located downstream from the diffuser (52). The problem with this device is that the velocity of the conveying gas stream (56) leaving the nozzle (51) can only vary within a relatively narrow range since otherwise clogging occurs in the suction chamber (53). In order to prevent deposits of solids in the suction chamber (53), a fluidizing gas (54) can

be blown into the suction chamber (53) from below through a screen. The attainable solids loading of the conveying-gas stream (56), the counter-pressure that can be overcome in the chamber downstream from the diffuser (52) and the gas outlet velocity from the diffuser (52) are all relatively low. Consequently, this system likewise entails few advantages in terms of solving the problem in question, namely, feeding dusts into the fluidized particle layer of fluidized-bed reactors and distributing them thoroughly in the layer.

DE-A 197 22 570 discloses a process and a reactor of the type mentioned above. This German patent suggests that gas streams be blown into fluidized particle layers of a fluidized-bed reactor by means of transversal injection at supersonic speed in order to avoid problems caused by local concentration differences. The suggestion is also made to blow gas-solids mixtures into the fluidized particle layer in this manner as well.

However, the technical realization of this suggestion is difficult since, in order to generate a gas jet with supersonic speed, the gas upstream from the supersonic nozzle (hereinafter also referred to as "Laval nozzle") has to be at a pressure of several bar and both the compression of gases containing dust as well as the feeding of dust into a compressed gas stream are extremely problematic. Therefore, with the known process, it is only possible to achieve weak solids loading of the gas stream at a relatively small depth of penetration into the fluidized particle layer.

The fluidized-bed reactor known from DE-A 197 22 570 has a slotted grate above which the fluidized particle layer is generated. This layer is surrounded by a reactor jacket in which Laval nozzles are installed. The Laval nozzles serve to accelerate to supersonic speed a solids-loaded gas stream flowing crosswise (transversally) to the main flow direction of the fluidizing gas towards the fluidized particle layer.

The invention has the objective of creating a simple and economical process with which a gas stream with high solids loading can be transversally blown with great penetration

depth into the fluidized particle layer for purposes of carrying out a reaction in a fluidized particle layer. Furthermore, the invention has the object of providing a simple and safe reactor to carry out the process.

When it comes to the process, this objective is achieved by the above-mentioned process in that, after leaving the supersonic nozzle, the propellant-gas stream is loaded with the solids powder and blown into the fluidized particle layer through a diffuser positioned across from the supersonic nozzle.

The propellant-gas stream is subsequently accelerated to supersonic speed by the supersonic nozzle and only then it is loaded with the solids powder. From a technical standpoint, compressing and accelerating the unloaded propellant-gas stream do not pose any particular difficulties. As a result of the high flow velocity of the propellant-gas stream, negative pressure is established in the area downstream from the supersonic nozzle. This effect is further enhanced by the diffuser so that it becomes easier to load the propellant-gas stream in the area upstream from the diffuser since the negative pressure causes the solids powder to be sucked into the propellant-gas stream. Therefore, the above-mentioned difficulties encountered with the compression of gases containing dust and with the feed of dust into a compressed gas stream do not occur in the case of the process according to the invention.

Since the propellant-gas stream leaves the supersonic nozzle (Laval nozzle) and enters the diffuser at supersonic speed, the solids powder can be conveyed from a chamber having a lower pressure into the reactor, where a higher pressure prevails. The process according to the invention allows a high solids loading of the propellant-gas stream, a constant suction rate and a high gas outlet velocity from the diffuser and thus a great depth of penetration of the solids powder into the fluidized particle layer.

In the fluidized particle layer, the solids powder reacts with the fluidizing gas that has been fed in. The reaction can consist of a chemical reaction or of a mechanical or thermal

treatment. An example of this is the return of pulverized, solid raw materials into the fluidized particle layer of the fluidized-bed reactor in the production of $TiCl_4$, $ZrCl_4$ and $SiCl_4$, which have been discharged from the reactor by the stream of fluidizing gas.

The diffuser is a component commonly employed in flow technology, by means of which a stream having a high velocity and low pressure is converted into a stream having a lower velocity and higher pressure. The diffuser has a flow channel with an inlet opening and an outlet opening for the flow medium.

The propellant-gas stream enters the fluidized particle layer in the transversal direction relative to the main flow direction of the fluidizing gas. In this context, the expression "transversal" comprises the directions that are crosswise to the main flow direction.

Advantageously, the solids powder is held in a suction chamber, from which it is sucked by means of the propellant-gas stream. The suction chamber surrounds the gas outlet side of the supersonic nozzle; it can encompass the entire area between the supersonic nozzle and the diffuser and serve as a buffer storage for a larger volume of the solids powder, as a result of which uniform loading of the propellant-gas stream is ensured.

Another improvement consists of placing the solids powder in the suction chamber at an elevated pressure. This can be done, for instance, in that the solids powder is fed at an elevated pressure from locks into the suction chamber. This makes it possible to further increase the solids loading and the depth of penetration of the propellant-gas stream.

In a particularly simple mode of operation, the propellant gas is used as the fluidizing gas. Due to the fact that they have the same chemical composition, the propellant gas and the fluidizing gas have the same effect on the solids powder.

For purposes of carrying out the chlorination reaction, preference is given to the use of a propellant gas that contains chlorine and/or oxygen.

The process according to the invention exhibits its full effect when the propellant gas is accelerated to a supersonic speed corresponding to at least 1.2 Mach, preferably 1.3 Mach to 3 Mach.

It has been found to be especially advantageous to create a negative-pressure zone in the area of the gas-outlet opening of the diffuser.
This additional zone of reduced pressure causes an increase in the depth of penetration of the loaded propellant-gas stream into the fluidized particle layer. Particularly in the case of fluidized-bed reactors having a large diameter, this measure is an alternative to the configuration entailing several feed devices. In this context, it has proven its worth to create the negative-pressure zone by blowing a propellant-gas stream into a pressure-reducing device connected to the diffuser. This makes it possible to increase the solids loading of the propellant-gas stream by several orders of magnitude in comparison to conventional methods. For example, the solids loading of the propellant-gas stream in the process according to the invention can be two to three times higher than with the above-mentioned pneumatic conveyance of the solids using conventional injectors, without the occurrence of clogging.

In a preferred process variant, raw material dust discharged from the fluidized particle layer is employed as solids powder. The process according to the invention makes it possible to take the dust discharged from the reactor during the production or processing operation and return it to the reactor so as to improve the cost effectiveness of the process. Advantageously, the raw material dust is separated directly from the hot reaction gas in a dust separator and then returned to the reactor by means of the process according to the invention. However, it is also possible to first process these valuable raw materials; for example, the raw material powder, together with solid metal chlorides, can be separated from the reaction gas of a fluidized-bed reactor and, after the metal chlorides have been washed out, it can be dried and then blown into the fluidized-bed reactor.

In another likewise preferred process variant, primary particles are utilized as the solids powder. Here, either part or all of the primary particles that are to be treated in the fluidized particle layer are brought into the reactor by means of the process according to the invention. With this measure, which stands out for its cost effectiveness, the primary particles as well as the raw material dust discharged from the fluidized particle layer can be employed as the solids powder.

In another advantageous process variant, waste dust is used as the solids powder. This waste dust consists of raw materials in dust form that are generated in other processes. Examples to be cited here are washed and dried TiO_2 waste dusts that occur as insoluble residues in the digestion of titanium raw materials with sulfuric acid.

Preferably, the propellant-gas stream is blown into the lower half, preferentially into the lower quarter, of the fluidized particle layer. In this manner, the solids powder reaches a lower zone of the fluidized particle layer so as to ensure a sufficient retention time in there for the reaction or treatment. In order to increase the retention time, the fluidized particle layer can contain additional particles made of an inert material.

For purposes of achieving a high solids loading of the propellant-gas stream, the distance between the supersonic nozzle and the diffuser is set in such a way that the pressure in the suction chamber is minimal.

Regarding the reactor used to carry out this process, the above-mentioned objective is achieved by the above-mentioned device according to the invention in that, seen in the direction of flow of the propellant-gas stream, a diffuser lies across from the supersonic nozzle and in that a suction chamber that serves to feed in a solids powder is provided between the supersonic nozzle and the diffuser.

The supersonic nozzle, diffuser and suction chamber are all components of a feed device that serves to feed a solids powder into the reactor. By means of the supersonic nozzle,

the propellant-gas stream is accelerated to supersonic speed in the direction of the diffuser. Between the supersonic nozzle and the diffuser, there is a suction chamber that serves to supply a solids powder. In its simplest version, the supersonic nozzle extends into the suction chamber. An outlet of a collecting tank or a line for solids powder that ends between the supersonic nozzle and the diffuser also works as a suction chamber as defined here. Solids powder is drawn from the suction chamber into the propellant-gas stream. The suction chamber can serve as a buffer storage for a larger volume of the solids powder, so that uniform loading of the propellant-gas stream and thus safe operation of the device are ensured.
Concerning process details and terminology, reference is hereby made to the explanations above regarding the process according to the invention.

The reactor can be fitted with several such feed devices for purposes of feeding in a solids powder.

The diffuser is a component commonly employed in flow technology, by means of which a stream having a high velocity and low pressure is converted into a stream having a lower velocity and higher pressure. The diffuser has a flow channel with an inlet opening and an outlet opening for the flow medium. A particularly simple and effective reactor is one in which the diffuser is in the form of a Venturi tube.

Another improvement of the reactor is attained in that the diffuser has a gas-outlet opening that is in functional connection with a pressure-reducing device. A negative pressure is generated opening by means of the pressure-reducing device in the area of the gas-outlet. This additional negative pressure results in a greater penetration depth of the loaded propellant-gas stream into the fluidizing particle layer. Particularly in the case of fluidized-bed reactors having a large diameter, this measure is an alternative to the configuration with several feed devices.

A pressure-reducing device designed in the form of an annular gap or a spiral-jet nozzle has proven to be especially well-suited for this purpose. One feed device can also be fitted with several such pressure-reducing devices. The pressure-reducing device makes it possible to increase the solids loading of the propellant-gas stream by several orders of magnitude in comparison to conventional methods.

Preferably, the supersonic nozzle can be moved towards the diffuser and in the opposite direction. As a result, it is very easy to set the distance between the supersonic nozzle and the diffuser in such a way that it has an effect on the negative pressure in the area of the outlet of the suction chamber and thus on the suction capacity of the propellant-gas stream.

The process and the reactor according to the invention will be elaborated upon below with reference to embodiments and a drawing. Sectional depictions in the drawing show the following in detail:

Figure 1 – a first embodiment of a feed device according to the invention for feeding a particle stream into a reactor, in a side view;

Figure 2 – another embodiment of such a feed device with a ring nozzle provided on the outlet side of the diffuser, in a side view;

Figure 3 – a diffuser provided with a spiral-jet nozzle, in a side view,

Figure 4 – a cross section through the diffuser according to Figure 3 along line A-A, in a top view; and

Figure 5 – a feed device for feeding a particle stream into a reactor according to the state of the art, in a side view.

The feed device shown in Figure 1 serves to feed solids dust into a fluidized-bed reactor, The feed device comprises a Laval nozzle **4**, a diffuser **2** in the form of a Venturi tube, and a suction chamber **3**. The Laval nozzle **4** opens into the suction chamber **3**. The diffuser **2** lies coaxially opposite from the Laval nozzle **4** in the suction chamber **3**. By means of the Laval nozzle **4**, propellant gas, which is symbolized by the directional arrow **11**, is accelerated to supersonic speed. The propellant gas **11** passes through the suction chamber **3** in the form of a propellant-gas stream. Solids dust that is to be fed into the fluidized-bed reactor is kept ready in the suction chamber **3**. The solids dust is conveyed to the suction chamber **3** via the feed opening **5**, as indicated by the directional arrow **12**. The propellant-gas stream draws the solids dust from the suction chamber **3** and then reaches the diffuser **2**, from where, in the form of a gas jet **13** containing solids, the stream enters the fluidized-bed reactor (not shown in the figure). Depending on the operating parameters of the feed device according to Figure 1, different levels of internal pressure are established in the area of the suction chamber **3**. In order to keep this internal pressure as low as possible, the distance between the Laval nozzle **4** and the diffuser **2** can be set by appropriately sliding the Laval nozzle **4** in the direction of the diffuser **2**, as indicated by the arrow **9**. The feed device is flanged from the outside onto the reactor jacket in the area of the lower quarter of the fluidized particle layer of the fluidized-bed reactor. For purposes of fluidizing the solids dust, the suction chamber **3** is provided with a gas inlet for a purging gas **14**.

In the depiction shown in Figure 2, the same reference numerals are used as in Figure 1 to designate identical or equivalent components of the feed device.

In addition, the feed device according to Figure 2 is provided on the outlet side facing the diffuser **2** with a pressure-reducing device in the form of an annular-gap nozzle **6**. This annular-gap nozzle **6** is fitted with a gas inlet through which an additional propellant-gas stream **15** is fed into the annular-gap nozzle **6**. In the area of the gas outlet of the diffuser **2**, the propellant-gas stream **15** generates a pressure drop that accelerates the gas jet **13**

leaving the diffuser **2**, thus bringing about a greater depth of penetration into the fluidized particle layer.

A similar effect is caused by a spiral-jet nozzle **7** flanged onto the diffuser **2** in the area of the gas outlet, as shown in Figures 3 and 4. The propellant-gas stream **15** is swirled by the spiral-jet nozzle **7**, which extends tangentially. In order to swirl the propellant-gas stream **15**, it is also possible to employ guide vanes as an alternative to the spiral-jet nozzle **7**.

From pneumatic conveying technology, injectors are known that consist of a suction chamber with a solids-feed opening, of a usually conical nozzle and of a diffuser arranged coaxially thereto.
These injectors can be deployed to convey fine-particle solids from a chamber having a lower pressure and located above the inlet opening into a chamber having a higher pressure and located downstream from the diffuser. The problem with this device is that the velocity of the conveying stream of the propellant gas leaving the nozzle can only be varied within a relatively narrow range since otherwise clogging occurs in the suction chamber. The attainable solids loading of the conveying-gas stream, the counter-pressure that can be overcome in the chamber downstream from the diffuser as well as the gas outlet velocity from the diffuser are all relatively low. Consequently, this system likewise entails few advantages in terms of solving the problem in question, namely, feeding dusts into the fluidized particle layer of fluidized-bed reactors and distributing them thoroughly in the layer.

The feed device shown in Figure 5 in the form of an injector known from the state of the art is described in greater detail above.

Below, the process according to the invention will be explained in greater detail with reference to an embodiment and to a comparative example with reference to the devices depicted in Figures 1 and 5:

Example 1 (Comparative Example)

In a fluidized-bed reactor, an injector according to Figure 5 was mounted on a fitting 30 cm above the inflow tray, as is commonly done in pneumatic conveying technology for purposes of conveying solids. The injector was flanged with the gas-outlet opening of the diffuser **52** onto the reactor wall. The diffuser **52** was in the form of a Venturi tube. The gas-outlet opening of the conical nozzle **51** had a diameter of 11 mm. Oxygen whose pressure had been reduced to 1.7 bar was employed as the propellant gas **56**. The optimal distance between the nozzle **51** and the diffuser **52** had been determined in advance by passing pulverized ore through a pipeline. The pressure generated by the fluidized layer in the fluidized-bed reactor pulsed between 1.15 bar and 1.20 bar in the area of the gas-inlet fitting. The ore dust was metered into the suction chamber **53** via a rotary-vane feeder whose rotational speed was controlled. At a propellant-gas pressure of 1.7 bar, 100 m^3 of O_2/h flowed through the nozzle **51** and the diffuser **52** into the reactor. In order to prevent deposits of solids in the suction chamber **53**, 5 m^3 of O_2/h as the fluidizing gas **54** were blown into the suction chamber **53** from below through a screen. Under these conditions, it was not possible to establish a uniform negative pressure in the suction chamber **53**. When more than 400 kg of ore dust/h were metered in, the suction chamber became clogged. Uniform metering was already no longer possible above 250 kg/h. The fraction of ore in the ore-coke dust mixture discharged from the reactor together with the reaction gases was greatly elevated, even at a metering rate of 200 kg of ore dust/h (all volume figures are in m^3 under normal conditions, all pressure figures refer to absolute pressure).

Example 2

Instead of the injector used in Example 1, the feed device depicted in Figure 1 was flanged onto the fluidized-bed reactor. Therefore, from a design standpoint, the conical nozzle **51** (Figure 5) was essentially replaced by a Laval nozzle (Figure 1).

The Laval nozzle **4** was dimensioned and manufactured in such a way that it generated a propellant-gas speed of 1.5 Mach at an oxygen admission pressure of 3.9 bar and at 200 m^3/h of O_2. The pneumatic conveying line was used to optimize the distance between the Laval nozzle **4** and the diffuser **2**. With this configuration and the cited process parameters, it was possible to feed in 2.5 t/h of ore dust into the reactor without the occurrence of excess pressure and clogging with solids in the suction chamber **3**. At a dust feed rate of up to 600 kg/h, the composition of the dust discharged from the reactor only changed negligibly. Between 80% and 90% of the dust blown into the reactor had reacted.

Patent Claims

1. A process to carry out a reaction between gaseous and solid reactants in a fluidized particle layer, whereby a fluidizing gas flows through a loose bed of primary particles, as a result of which the bed rises while forming the fluidized particle layer and then reacts with the primary particles, and whereby a stream of a propellant gas loaded with solids powder and accelerated to supersonic speed by means of a supersonic nozzle is blown into the fluidized particle layer transversally to the main flow direction of the fluidizing gas, characterized in that, after leaving the supersonic nozzle (4), the propellant-gas stream (11) is loaded with the solids powder and blown into the fluidized particle layer via a diffuser (2) positioned across from the supersonic nozzle (4).

2. The process according to Claim 1, characterized in that the solids powder is held in a suction chamber (3), from which it is sucked by means of the propellant-gas stream (11).

3. The process according to Claim 2, characterized in that the solids powder is held in a suction chamber (3) at an elevated pressure.

4. The process according to one of the preceding claims, characterized in that the fluidizing gas is used as the propellant gas.

5. The process according to one of the preceding claims, characterized in that the propellant gas contains chlorine and/or oxygen.

6. The process according to one of the preceding claims, characterized in that the propellant gas is accelerated to a supersonic speed corresponding to at least 1.2 Mach, preferably 1.3 Mach to 3 Mach.

7. The process according to one of the preceding claims, characterized in that the diffuser (2) has a gas-outlet opening and in that a zone of negative pressure is created in the area of the gas-outlet opening.

8. The process according to Claim 7, characterized in that the negative-pressure zone is generated by blowing a propellant-gas stream (15) into a pressure-reducing device (6; 7) connected to the diffuser (2).

9. The process according to one of the preceding claims, characterized in that the raw material dust discharged from the fluidized particle layer is used as the solids powder.

10. The process according to one of the preceding claims, characterized in that primary particles are used as the solids powder.

11. The process according to one of the preceding claims, characterized in that waste dust is used as the solids powder.

12. The process according to one of the preceding claims, characterized in that the propellant-gas stream (11) is blown into the lower half, preferably into the lower quarter, of the fluidized particle layer.

13. The process according to Claim 2 or 3, characterized in that the distance between the supersonic nozzle (4) and the diffuser (2) is set in such a way that the pressure in the suction chamber (3) is minimal.

14. A reactor to carry out a reaction in a fluidized particle layer, said reactor having a feed tray through which a fluidizing gas is fed into a bed of primary particles located above the inflow tray in order to generate the fluidized particle layer, and having a reactor wall that surrounds the fluidized particle layer, into which wall at least one feed device is inserted above the inflow tray, said device comprising a supersonic nozzle by means of which a propellant gas is accelerated to supersonic speed, thereby generating a propellant-gas stream directed transversally to the main flow direction of the fluidizing gas, characterized in that, seen in the direction of flow of the propellant-gas stream (11), the supersonic nozzle (4) lies across from a diffuser (2) and in that a suction chamber (3) that serves to feed in a solids powder is provided between the supersonic nozzle (4) and the diffuser (3).

15. The reactor according to Claim 14, characterized in that the diffuser (2) is designed in the form of a Venturi tube.

16. The reactor according to Claim 14 or 15, characterized in that the diffuser (2) has a gas-outlet opening that is in functional connection with a pressure-reducing device (6; 7).

17. The reactor according to Claim 16, characterized in that the pressure-reducing device is designed in the form of an annular gap (6) or a spiral-jet nozzle (7).

18. The reactor according to one of Claims 14 through 17, characterized in that the supersonic nozzle (4) can be moved in the direction indicated by (9) towards the diffuser (2) and in the opposite direction.

Process and device to carry out a reaction between gaseous and solid reactants in a fluidized particle layer

In order to carry out a reaction between gaseous and solid reactants in a fluidized particle layer, normally a fluidizing gas flows through a loose bed of primary particles, as a result of which the bed rises while forming the fluidized particle layer and then reacts with the primary particles. It is also a known process to blow a stream of a propellant gas (11) loaded with solids powder and accelerated to supersonic speed by means of a supersonic nozzle transversally into the main flow direction of the fluidizing gas into the fluidized particle layer. In order to increase the solids loading and the depth of penetration of the propellant-gas stream (11), it is proposed that, after leaving the supersonic nozzle (4), the propellant-gas stream (11) is loaded with the solids powder and blown into the fluidized particle layer via a diffuser (2) positioned across from the supersonic nozzle (4). A simple and safe reactor to carry out the process is characterized by a supersonic nozzle (4) which, seen in the direction of flow of the propellant-gas stream (11), lies across from a diffuser (2), whereby a suction chamber (3) that serves to feed in solids powder is provided between the supersonic nozzle (4) and the diffuser (2).

(Figure 1)

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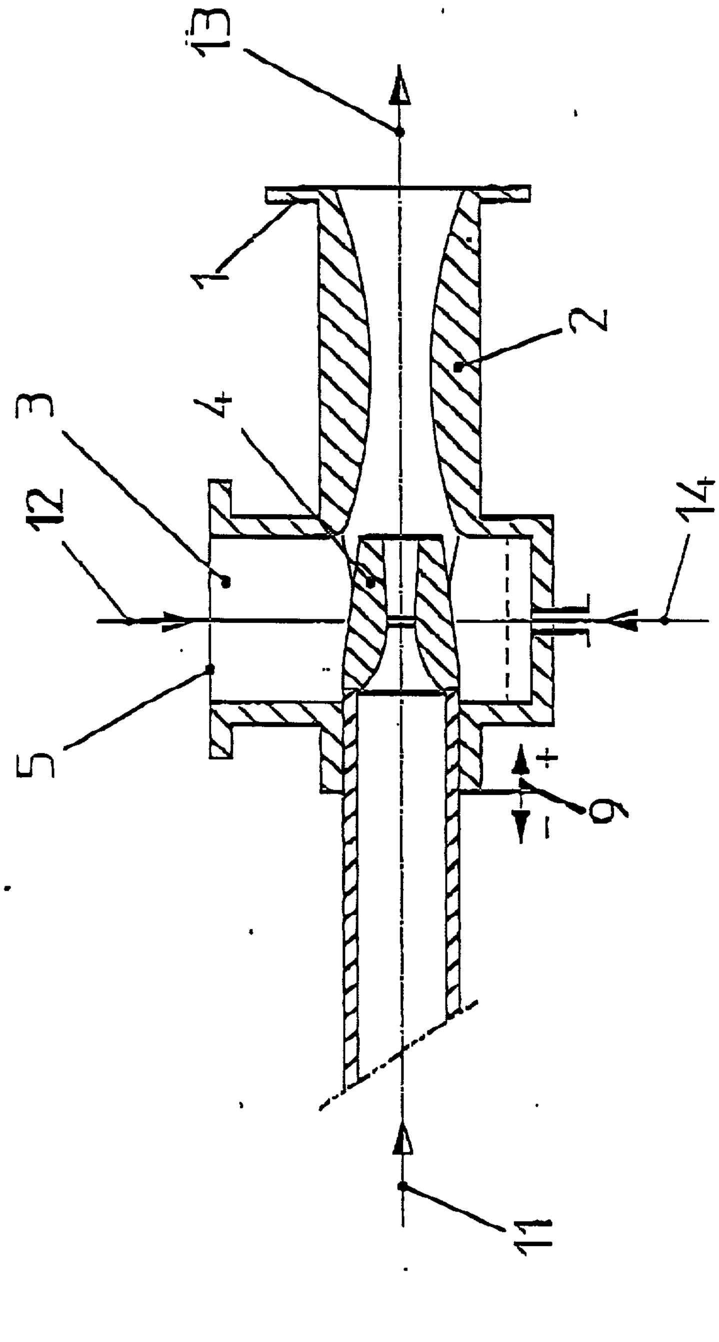

Fig. 1

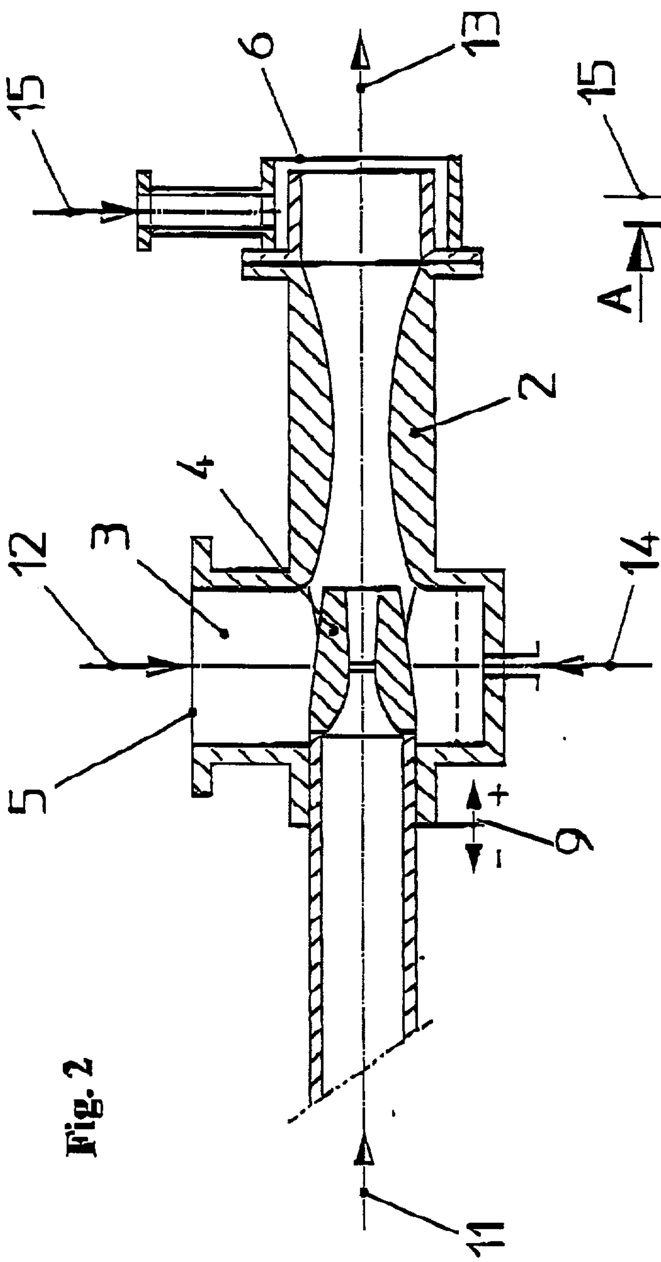

Fig. 2

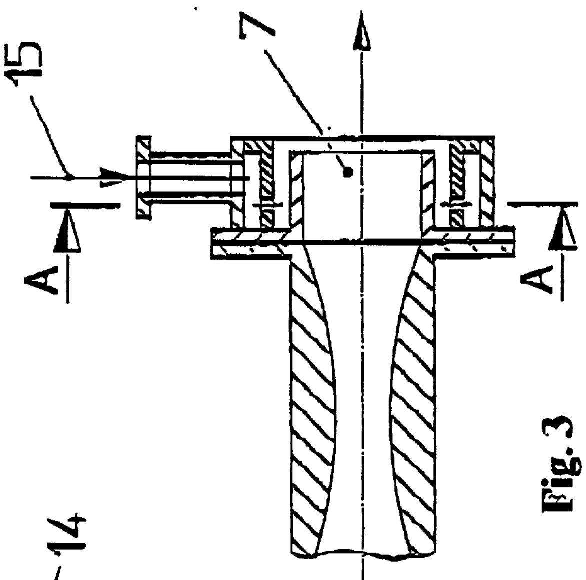

Fig. 3

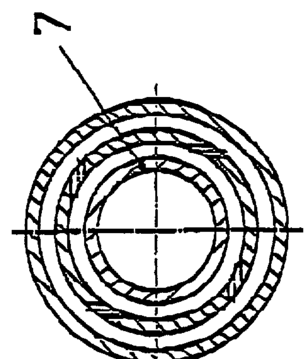

Fig. 4

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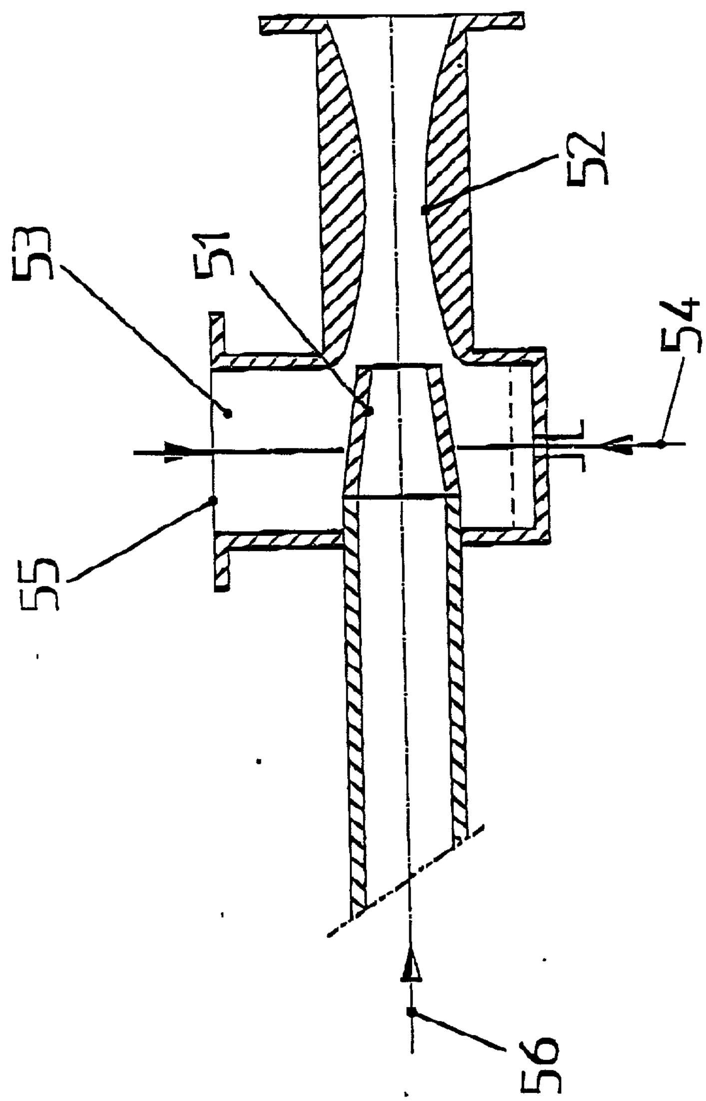

Fig. 5

Declaration and Power of Attorney for Patent Application

Erklärung für Patentanmeldungen mit Vollmacht

German Language Declaration

Als nachstehend benannter Erfinder erkläre ich hiermit an Eides Statt:

daß mein Wohnsitz, meine Postanschrift und meine Staatsangehörigkeit den im nachstehenden nach meinem Namen aufgeführten Angaben entsprechen, daß ich nach bestem Wissen der ursprüngliche, erste und alleinige Erfinder (falls nachstehend nur ein Name angegeben ist) oder ein ursprünglicher, erster und Miterfinder (falls nachstehend mehrere Namen aufgeführt sind) des Gegenstandes bin, für den dieser Antrag gestellt wird und für den ein Patent für die Erfindung mit folgendem Titel beantragt wird.

____

____

deren Beschreibung hier beigefügt ist, es sei denn (in diesem Falle Zutreffendes bitte ankreuzen), diese Erfindung

☐ wurde angemeldet am __________ unter der US-Anmeldenummer oder unter der Internationalen Anmeldenummer im Rahmen des Vertrags über die Zusammenarbeit auf dem Gebiet des Patentwesens (PCT)

______________ und am

______________ abgeändert (falls zutreffend).

Ich bestätige hiermit, daß ich den Inhalt der oben angegebenen Patentanmeldung, einschließlich der Ansprüche, die eventuell durch einen oben erwähnten Zusatzantrag abgeändert wurde, durchgesehen und verstanden habe.

Ich erkenne meine Pflicht zur Offenbarung jeglicher Informationen an, die zur Prüfung der Patentfähigkeit in Einklang mit Title 37, Code of Federal Regulations, § 1.56 von Belang sind.

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

METHOD AND APPARATUS FOR INTRODUCING POWDERED RAW MATERIAL INTO A FLUIDIZED PARTICLE BED

the specification of which is attached hereto unless the following box is checked:

☐ was filed on June 11, 2001 as United States Application Number or PCT International Application Number 09/857,895

______________ and was amended on

______________ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

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Ich beanspruche hiermit ausländische Prioritätsvorteile gemäß Title 35, US-Code, § 119 (a)-(d), bzw. § 365(b) aller unten aufgeführten Auslandsanmeldungen für Patente oder Erfinderurkunden, oder § 365(a) aller PCT internationalen Anmeldungen, welche wenigstens ein Land ausser den Vereinigten Staaten von Amerika benennen, und habe nachstehend durch ankreuzen sämtliche Auslandsanmeldungen für Patente bzw. Erfinderurkunden oder PCT internationale Anmeldungen angegeben, deren Anmeldetag dem der Anmeldung, für welche Priorität beansprucht wird, vorangeht.

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Prior Foreign Applications
(Frühere ausländische Anmeldungen)

Priority Claimed
Priorität beansprucht

(Number) (Nummer)	(Country) (Land)	(Day/Month/Year Filed) (Tag/Monat/Jahr der Anmeldung)	YES	NO
PCT/EP 00/09789	PCT	OCTOBER 6, 2000	X	☐
			X	☐

Ich beanspruche hiermit Prioritätsvorteile unter Title 35, US-Code, § 119(e) aller US-Hilfsanmeldungen wie unten aufgezählt.

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(Application No.) (Aktenzeichen) — (Filing Date) (Anmeldetag)

(Application No.) (Aktenzeichen) — (Filing Date) (Anmeldetag)

Ich beanspruche hiermit die mir unter Title 35, US-Code, § 120 zustehenden Vorteile aller unten aufgeführten US-Patentanmeldungen bzw. § 365(c) aller PCT internationalen Anmeldungen, welche die Vereinigten Staaten von Amerika benennen, und erkenne, insofern der Gegenstand eines jeden früheren Anspruchs dieser Patentanmeldung nicht in einer US-Patentanmeldung, bzw. PCT internationalen Anmeldung in einer gemäß dem ersten Absatz von Title 35, US-Code, § 112 vorgeschriebenen Art und Weise offenbart wurde, meine Pflicht zur Offenbarung jeglicher Informationen an, die zur Prüfung der Patentfähigkeit in Einklang mit Title 37, Code of Federal Regulations, § 1.56 von Belang sind und die im Zeitraum zwischen dem Anmeldetag der früheren Patentanmeldung und dem nationalen oder im Rahmen des Vertrags über die Zusammenarbeit auf dem Gebiet des Patentwesen (PCT) gültigen internationalen Anmeldetags bekannt geworden sind.

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(Application No.) (Aktenzeichen) — (Filing Date) (Anmeldetag) — (Status) (patented, pending, abandoned) (Status) (patentiert, schwebend, aufgegeben)

(Application No.) (Aktenzeichen) — (Filing Date) (Anmeldetag) — (Status) (patented, pending, abandoned) (Status) (patentiert, schwebend, aufgegeben)

Ich erkläre hiermit, daß alle in der vorliegenden Erklärung von mir gemachten Angaben nach bestem Wissen und Gewissen der Wahrheit entsprechen, und ferner daß ich diese eidesstattliche Erklärung in Kenntnis dessen ablege, daß wissentlich und vorsätzlich falsche Angaben oder dergleichen gemäß § 1001, Title 18 des US-Code strafbar sind und mit Geldstrafe und/oder Gefängnis bestraft werden können und daß derartige wissentlich und vorsätzlich falsche Angaben die Rechtswirksamkeit der vorliegenden Patentanmeldung oder eines aufgrund deren erteilten Patentes gefährden können.

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MG 2280

German Language Declaration

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POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: *Rudolf E. Hutz, Reg. No. 22,397; Harold Pezzner, Reg. No. 22,112; John D. Fairchild, Reg. No. 19,756; Richard M. Beck, Reg. No. 22,580; Paul E. Crawford, Reg. No. 24,397; Thomas M. Meshbesher, Reg. No. 25,083; Patricia Smink Rogowski, Reg. No. 33,791; Robert G. McMorrow, Jr., Reg. No. 30,962; Ashely I. Pezzner, Reg. No. 35,646; William E. McShane, Reg. No. 32,707; Mary W. Bourke, Reg. No. 30,982; Gerard M. O'Rourke, Reg. No. 39,79, all of P. O. Box 2207, Wilmington, Delaware 19899-2007, my attorneys with full power of substitution and revocation.*

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(Supply similar information and signature for third and subsequent joint inventors.)